Plasmonic Gap Modes Enhanced Optical Harmonic Generation in Metal Film-coupled Nanoparticles Towards the Quantum Tunneling Regime

Dangyuan Lei

Department of Applied Physics, The Hong Kong Polytechnic University, Hong Kong

Email of Presenting Author: dylei@polyu.edu.hk

The rapid development in nanofabrication and characterization has driven the miniaturization of plasmonic nanostructures towards subnanometer - and even atomic-scale feature sizes. At such small length scales, atomic interaction and quantum repulsion within an isolated metal nanostructure significantly modify the macroscopic dielectric response of metal such that the spatial nonlocality effect has to be conisdered as necessary correction to the conventional Drude model. In particular, upon formation of a nanoparticle dimer or particle-on-film junction, both separated by subnanometer gaps, the quantum mechanical effects including the spill out of electrons and electron tunneling across the junction have to be taken into account for precisely describing their linear and nonlinear plasmonic properties. Though these effects can significantly reduce the plasmon-induced field enhancement at the gaps of the coupled systems, recent theories show that the electron tunneling across the gaps strongly enhances the effective optical nonlinearities of metals that in turn benefits the generation of optical high harmonics. In this talk, I will show our recent experimental investigation on the nonlinear optical response of metal film-coupled nanoparticle monomers and dimers as a function of the gap distance. Single-particle spectroscopic measurements indicate that this hybrid nanostructure can simultaneously enhance several nonlinear optical processes, including the second harmonic generation (SHG), two-photon induced luminescence, and hyper-Raman scattering. The junction gap distance dependent SH signal is expected to unambiguously reveal the critical transition from the classical plasmon-enhanced SHG to the electron tunneling induced additional nonlinear coefficients in metals.